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MODELING EMISSIONS BY CHEMICAL KINETICS TO A GENSET WITH BLENDS OF DIESEL-BIODIESEL

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Abstract. *In recent years, biofuels have been identified as alternatives and possible solutions to the problems caused by the use of fossil fuels in energy production. Oils from different sources (fossil fuels, biodiesel from vegetable sources and diesel from microalgae i.e.) have been the subject of study by the scientific community. However, despite being sustainable and biodegradable, there are indications that engines fueled with biodiesel blends emit pollutants such as nitrogen oxides in greater quantities than when fueled by fossil diesel. The main objective was to develop a computationally lightweight model, from the point of view of chemical kinetics, but with the support of well-collected experimental data, through adjustment of mathematical models and experimental validation, thus producing accurate results. To determine the kinetic constants, the problem was faced as a situation of consumption and production, and to describe the analyzed scenario, seven species conservation reactions were proposed in this work, with that 15 kinetic constants to be determined. The Nelder-Mead Simplex Algorithm was selected to minimize errors between the model and experimental data. And, a computational code based on the FORTRAN® language was written. The validation of the proposed model was carried out by an experiment with a motor generator set (Genset) from Maquigeral model 12W, with a 44kW capacity; the diesel engine installed is a MWM D229/4 with natural aspiration and is coupled to the WEG GTA 201 AIHV generator. The Genset is installed in the Sustainable Energy Research and Development Center (NPDEAS), and an output power (energy load) of 5kW was applied, through the energy consumption of the building, at a constant rotation speed of 1800 rpm. With B11 fuel blends (petroleum diesel plus 11% biodiesel), B25, B50, B75 and B100. From this, the emissions of nitrogen and carbon oxides were measured. By evaluating the results between loads, the researchers developed a model capable of demonstrating how the emissions productions are influenced by the increase in the engine operating temperature. Moreover, considerably relevant results were obtained from the mathematical model for CO_2 emissions, showing an error of 1% when compared to the experimental data.*
Keywords: *Emissions, Combustion, Model, ICO engine, GHG Emissions, Nelder-Mead method, Biofuels, Diesel, Biodiesel*

1. INTRODUCTION

Technological progress is closely linked to the rise in energy consumption. According to Ritchie and Roser (2014) there is substantial evidence supporting this relationship. The first set of data consists of a historical series depicting global primary energy consumption, since 1800, which coincides with the advent of the Industrial Revolution. During this period, energy consumption skyrocketed from less than 10,000 TWh per year to over 140,000 TWh per year. The most significant

surge occurred around the 1950s when annual consumption reached approximately 30,000 TWh. In other words, energy consumption tripled in 150 years (1800 - 1950) and then more than quadrupled in the last 70 years (1950 - 2018). This means that the annual increase in consumption during the latter period was almost 12 times higher (around 130 TWh/year from 1800 to 1950 vs 1500 TWh/year from 1950 to 2018).

Furthermore, the study reveals that the surge in energy usage is predominantly driven by non-renewable and polluting sources such as oil, coal, and natural gas. Consequently, our energy consumption is increasing, and there seems to be a preference for traditional sources over renewables (British Pretoleum, 2004) and (Jensen, 1985)

The second dataset sheds light on the current energy consumption in relation to GDP, comparing well-developed countries with less developed ones. For instance, countries like Australia and Sweden, with a GDP per capita of around $45,000, exhibit energy consumption levels of 60,000 kWh per capita. On the other hand, countries like Chile and Turkey, with a GDP per capita of approximately $25,000, have an energy usage of around 20,000 kWh per capita. The majority of high energy-consuming countries are located in Europe, while sub-Saharan African countries dominate the lower end of the consumption spectrum (Our World In Data, 2015; and Chontanawat et al., 2008).

Consequently, energy consumption will continue to grow as long as society strives for societal improvement, not only in technological aspects. (ANP, 2016; and Atabani et al., 2012). However, the increase in energy usage does not come without consequences. It is widely recognized that most energy production methods have adverse environmental impacts. Even the most environmentally friendly sources, such as wind and hydroelectric power, are associated with significant issues like animal fatalities caused by wind turbines and the disruption of soil ecosystems due to the construction of hydroelectric power plants (Berchin, 2015; Agarwal, 2016; and Saidur, 2011).

Recent literature includes studies and models focused on emissions production from diesel-biodiesel driven engines. These models employ various methods and approaches to describe and predict the relationship between emissions and factors such as fuel blend variation and engine parameters. Some commonly used techniques include the KIVA model (Li et al., 2013; An et al., 2014; and Aldhaidhawi et al., 2019a), as well as similar Computational Fluid Dynamics (CFD) software (Aldhaidhawi et al., 2017b). Additionally, empirical models have been developed (Anwar et al., 2020; Mejiá et al., 2019; and Simsek and Uslu, 2020). However, these models have certain limitations and gaps in their investigation. Empirical models rely solely on empirical data sets without incorporating analytical or chemical modeling. On the other hand, CFD models often have limited experimental data, particularly concerning fuel blend variation. They may only present comparisons between two or three different blends or a restricted range of analyzed emissions.

This study introduces a novel chemical model based on production and consumption equations. The model is designed to address these gaps and is adjusted and validated using experimental data encompassing a broader fuel blend range, ranging from B11 (almost pure diesel) to B100 (pure biodiesel). Consequently, the proposed model covers a wide spectrum of possible fuel mixtures. Furthermore, this study examines and analyzes three distinct emission species simultaneously: carbon monoxide, carbon dioxide, and nitrogen oxides (NOx). Notably, there is currently no work in recent literature that presents a theoretical-empirical mixed model with extensive experimental validation, encompassing a wide range of different emissions as comprehensively as the model proposed in this study.

2. METHODOLOGY

2.1 Kinetical Model

To build the mathematical model of the emissions' production, a kinetic approach was chosen. It was decided to face the problem as a consumption–production situation. So, then it has a combustion process and an extra process of production of nitrogen oxides. The model consists in equations referenced from (1) to (7):

1. An equation for the consumption of (D) diesel ($C_{12}H_{26}$);
2. An equation for the consumption of (BD) biodiesel ($C_{20}H_{36}O_2$);
3. An equation for the consumption of oxygen (O_2);
4. An equation for the consumption of nitrogen (N_2);
5. An equation for the production/consumption of (CO);
6. An equation for the production of (CO_2); and
7. An equation for the production of (NO_x).

$$\frac{d[\mathrm{D}]}{dt} = -k_1[\mathrm{D}][\mathrm{O}_2] \tag{1}$$

$$\frac{d[\mathrm{BD}]}{dt} = -k_2[\mathrm{BD}][\mathrm{O}_2] \tag{2}$$

$$\frac{d[O_2]}{dt} = -k_3[D][O_2] - k_4[BD][O_2] - k_5[CO][O_2] - k_6[N_2][O_2] \tag{3}$$

$$\frac{d[N_2]}{dt} = -k_7[N_2][O_2] \tag{4}$$

$$\frac{d[CO]}{dt} = k_8[D][O_2] + k_9[BD][O_2] - k_{10}[CO][O_2] \tag{5}$$

$$\frac{d[CO_2]}{dt} = k_{11}[D][O_2] + k_{12}[BD][O_2] + k_{13}[CO][O_2] \tag{6}$$

$$\frac{d[NO_x]}{dt} = k_{14}[N_2][O_2] + k_{15}[BD][O_2] \tag{7}$$

Where [D] is the concentration of diesel in the system, [BD] the concentration of biodiesel, [E] is the concentration of elements E={O_2, CO, CO_2, N_2, NO_x} and k_i for i={1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15} are the kinetics constants referent to that pair of elements' interactions. For example, in Eq. 4, k_7 is the kinetic constant linked to the interaction of N_2 and O_2 to consume, in this particular system, N_2. So, the constant will be an indicator of how intense these two compounds interact to consume N_2.

With these equations stated, the goal is to determine the values of the 15 kinetic constants presented on this simplified model. Five sets of data were chosed to serve as guidance for the adjustment of the model. These data were collected by Matiolo et al. (2020) in an MWM D229 diesel engine. The chosen data were five different diesel-biodiesel mixtures all in one engine load, 5kW, which represents 11% of the engine maximum load capacity. The mixtures were 11% of biodiesel, 25%, 50%, 75%, and finally 100% of biodiesel. As the operational rotation of the referred diesel engine is known to be 1800 RPM, and the engine needs two cycles to complete a thermodynamic cycle, the fuel consumption was taken from kg/h to grams/cycle. In this way, the model will be set to estimate the mean emissions production per cycle of the engine thermodynamic process.

2.2 Numerical Method

To numerically solve the system, a Runge-Kutta-Fehlberg method (Kincaid and Chaney, 2002) was used. The initial conditions for the system are the main inputs of the model. Here, the measurement of fuel consumption in Matiolo et al. (2020) was the source for the initial fuel quantities. For the nitrogen and oxygen availability, the stoichiometric proportion for the combustion reaction was used. The reaction was modeled by Graciano et al. (2016) as indicated in the equation (8):

$$\begin{aligned} yC_\delta H_\zeta O_\xi + (1-y)\left\{xC_\alpha H_\beta + (1-x)\left[zCH_4 + (1-z)CO_2\right]\right\} + \lambda a\left(O_2 + 3{,}76N_2\right) \Rightarrow \\ \Rightarrow bCO_2 + cH_2O + 3{,}76\lambda aN_2 + (\lambda - 1)aO_2 \end{aligned} \tag{8}$$

where *y* represents the molar biodiesel fraction on the mixture, λ is the excessive air ratio (quotient between real and stoichiometric air-fuel ratios), δ, ζ and ξ are the numbers of carbon, hydrogen, and oxygen on the biodiesel molecule, respectively; α and β, the numbers of carbon and hydrogen on the diesel molecule; and *a*, *b* and *c*, the balanced coefficients in the combustion reaction. The excessive air ratio is where the engine load is taken into consideration and its values were measured in Matiolo et al. (2020) experiments. The composition of both biodiesel and diesel was simplified in the same manner as it was done in Graciano et al. (2016), being $C_{12}H_{36}O_2$ and $C_{12}H_{26}$, respectively.

Table 1: Relation between biodiesel's mass percentage and molar fraction in fuel mixture

Fuel mixture in mass (%)	Biodiesel Molar Fraction (γ)
B11	0,062821931
B25	0,153107465
B50	0,351648541
B75	0,619369708
B100	1,000000000

As in the combustion reaction equation, the molar fraction is used, and in Matiolo et al. (2020) experiments, the fuel mixture is done relative to fuel mass and the molecules of diesel and biodiesel have different molecular weights, a relation had to be done, so the coefficient *y* truly represents the desired mixture. For a B11 mixture, for example, the molar fraction *y* that will give the 11% in mass of biodiesel is around 0,0628. All the molar fractions relative to the mixtures are listed in Table 1.

2.3 Constants Obtainment and Error Minimization

The next and greatest challenge is to determine the values of the 15 kinetic constants presented on the mathematical model. For this, a Nelder-Mead Simplex Method (Luersen and Le Riche, 2004; Nelder and Mead, 1965; and Wright, 2012) was chosen to numerically minimize the error function between the theoretical value obtained by the model and the experimental data collected. O'Neill, 1971, wrote the algorithm that was implemented in FORTRAN® (O'Neill, 1971). In other words, the Nelder-Mead Method minimizes the squared error function, while this is defined by equation (9):

$$\sum_{j=1}^{6} W_j (c_{\exp,j} - c_{t,\mathrm{j}})^2 \tag{9}$$

in which c_t are the results of the kinetic model, and c_{exp} are the collected experimental data referent to the result *j*:

- 1 to produced CO;
- 2 to produced CO_2;
- 3 to produced NO_X;
- 4 to consumed diesel;
- 5 to consumed biodiesel; and
- 6 to mass conservation of the system.

The expected results are: The experimental data collected by Matiolo et al. (2020) for the emissions productions; the Diesel and Biodiesel to be completely consumed with the sum of all masses at the beginning (Fuel mixture, Nitrogen, and Oxygen) (Eq. 1, 2, 3 and 4), to be the same as in the end (produced emissions, remaining oxygen, nitrogen, and residual fuel) (Eq. 5, 6 and 7).

The vector W_j is a weight vector can control the impact of each error function in the final error, giving us a tool to point the algorithm efforts into a better results direction. This vector provides us with a tool so that can guide the results of the error function towards more coherent directions from a physical point of view.

If mass conservation errors are to be more influential, just increase the W_6, for example. In this way, we have a simple tool to help guide the algorithm towards a coherent solution.

The convergence point was determined by a fixed number of interactions. The best results were extracted after 20.000 interactions. After reaching the convergence point, the algorithm delivered the best kinetic constants available within the set of initial conditions.

3. Results and Discussion

After several attempts to properly balance the weight vector in order to achieve physically acceptable results, the optimal combination was discovered. This combination produced the set of kinetic constants after the previously set 20,000 interaction.

With the constants determined, it was possible to solve numerically the chemical model proposed by Equations from 1 to 7. The solution resulted in the concentration of each emission produced in every one of the five proposed scenarios, which were each one of the five fuel mixtures: B11, B25, B50, B75 and B100. The results combined are exposed in Figure 1.

In Figure 1, the first graph 1.a shows the comparison of the concentration of carbon monoxide ([CO]) in ppm measured by Matiolo et al. (2020) and the results obtained by the model proposed. The outline produced by the model's results seems to indicate a trend very close to the real concentrations. The best values found for this particular emission were in B50 and B75, being 3,37% and 8,80%. On the other hand, the B11 and B25 were outside the measurement uncertainty with 18,33% and 35,41% of error. The profile shows a slightly different behavior compared to experimental data, but the results are acceptable.

The second graph 1.b is the comparison between the experimental and calculated data of the carbon dioxide ([CO_2]) concentration measured in %IR. These results show an excellent estimated profile calculated by the model in all of them blends, presenting errors less than 2,63%.

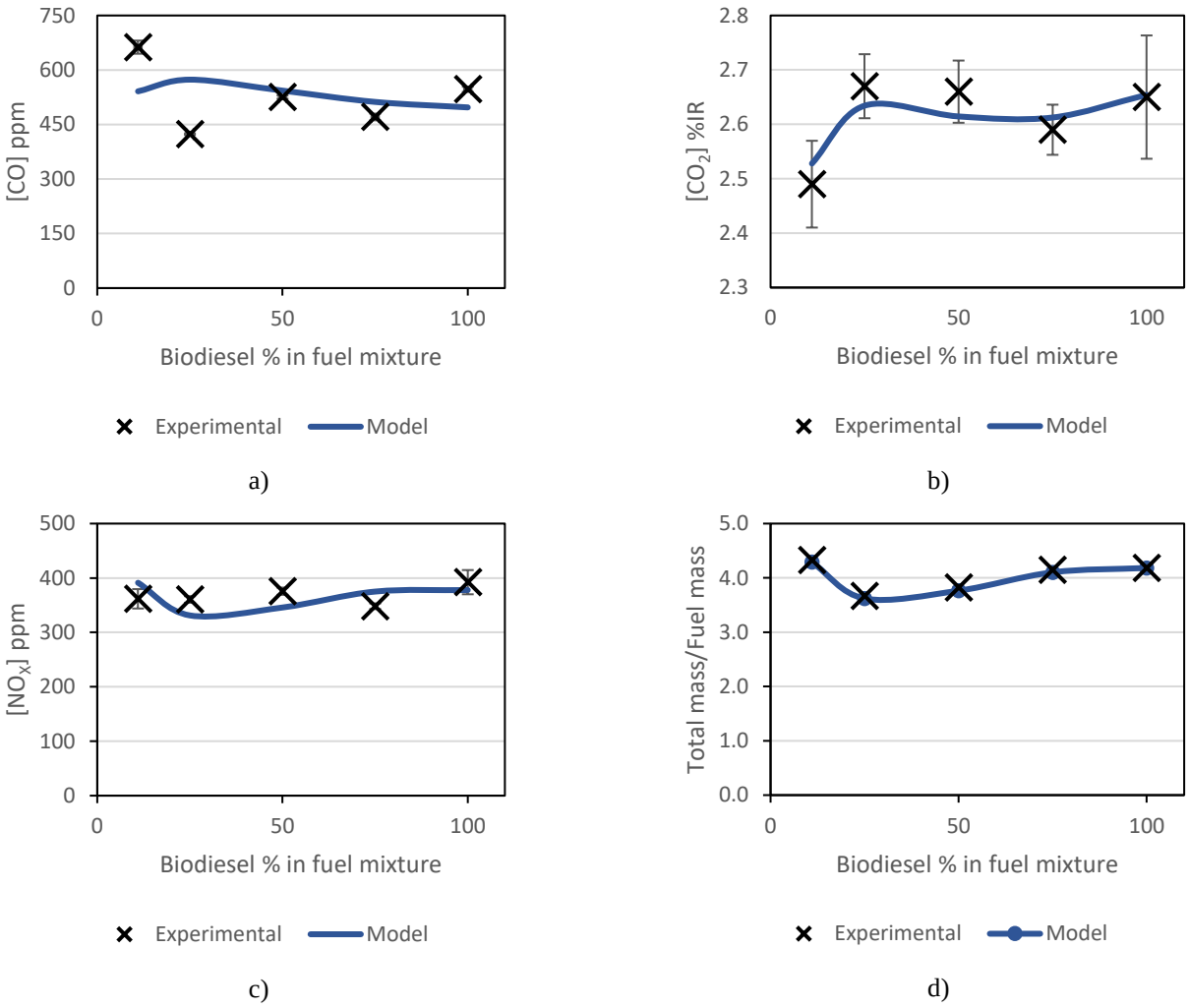

Figure 1: Emissions produced and mass conservation by different fuel mixtures. Experimental and modeled data comparison: a) CO (ppm) and b) CO_2 (%ir). C) NO_x (ppm) d) Dimensionless mass conservation

The third graph 1.c exposes the comparison between model results and experimental data of the nitrogen dioxides concentrations ([NO_x]). The most relevant errors are noted on B11 and B25, with modules of 8,2% and 8,1% respectively. Only the B100is inside the measurement uncertainty, with error is significantly minor, being only 3,7%. The profile shows a slightly different behavior compared to experimental data, but the results are acceptable. s described in the methodology; the applied load was one of the adjustment points of the fifteen kinetic constants that were presented in the model. Through the simplex or Nelder-Mead method, the adjustment of the CO, CO_2, and NOx production curves was achieved. The error obtained for each emission compared to the experimental data were 9,12%, 1,34% and 7,96%, respectively, accounting for a satisfactory total mean error of 7,76%. More than that, the production trend curves as a function of the fuel mixture, show an interesting behavior even in view of the complexity of the behavior observed through the analysis of the experimental data.

The trend curves generated through the model with the constants calculated by the Nelder-Mead method presented at the Figure 1. It is noted here that the behavior of the data does not have an obvious tendency towards the behavior of the fuel mixture, in addition to having an inherent complexity to the model, since the constants interact with the concentrations of the compounds. These often appear in more than one of the model's production/consumption equations.

The graph 1.d shows the dimensionless mass (total mass and fuel mass quotient) conservation. These results demonstrated no error near of 1%, bringing a physically strong argument in favor of the reliability of the model.

The errors obtained for each emission compared to the experimental data of CO, CO_2 and NO_X were 9,2%, 1,34% and 7,96%, respectively, accounting for a satisfactory total mean error of 7,76%. The average error of the results was found to be 4,90%, including the mass conservation. More than that, the production trend curves as a function of the fuel mixture,

show an interesting behavior even in view of the complexity of the behavior observed through the analysis of the experimental data.

4. FINAL CONSIDERATIONS

The results obtained by the model and method used show this the, provided curves of the emissions production would behave with the variation of the fuel mixture used to feed the engine. By evaluating the results between loads, it was obtained have a model capable of showing how the emissions productions are influenced by the increase in the engine operating temperature.

It is valuable noting that, despite the simplicity of the model, using only differential equations of production and consumption, a profile was obtained to characterize the production of three different emissions, correlated by the model. Even with the experimental data behaving without following a known trend or evolution profile along the diesel-biodiesel blends and in almost all the possibilities of blending biodiesel.

This work was conducted with the aim of advancing the knowledge and understanding of energy production processes, leading to an increased progression in human development while minimizing the impact on the environment in which the researchers operate. The ultimate goal is to move towards sustainable development.

A mathematical model is proposed based on species production and consumption equations with seven equations (diesel, biodiesel, oxygen and nitrogen; carbon monoxide, carbon dioxide and nitrogen oxides) and composed of fifteen constants, which integrates perfectly with the thermodynamic model of Graciano et al. (2016). This union allows us to evaluate different types of ICO engines, just by their geometric and operating parameters, creating a fast tool with a wide possibility of use in the evaluation of the emission productions of engines powered by fuel mixtures of any composition between B11 and B100.

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